

GEOCHEMICAL EFFECTS ON THE BEHAVIOR OF LLW RADIONUCLIDES IN SOIL/GROUNDWATER ENVIRONMENTS

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ABSTRACT

Assessing the migration potential of radionuclides leached from low-level radioactive waste (LLW) and decommissioning sites necessitates information on the effects of sorption and precipitation on the concentrations of dissolved radionuclides. Such an assessment requires that the geochemical processes of aqueous speciation, complexation, oxidation/reduction, and ion exchange be taken into account. The Pacific Northwest National Laboratory (PNNL) is providing technical support to the U.S. Nuclear Regulatory Commission (NRC) for defining the solubility and sorption behavior of radionuclides in soil/ground-water environments associated with engineered cementitious LLW disposal systems and decommissioning sites. Geochemical modeling is being used to predict solubility limits for radionuclides under geochemical conditions associated with these environments. The solubility limits are being used as maximum concentration limits in performance assessment calculations describing the release of contaminants from waste sources. Available data were compiled regarding the sorption potential of radionuclides onto "fresh" cement/concrete where the expected pH of the cement pore waters will equal to or exceed 10. Based on information gleaned from the literature, a list of preferred minimum distribution coefficients (K_d 's) was developed for these radionuclides. The K_d values are specific to the chemical environments associated with the evolution of the compositions of cement/concrete pore waters.

BACKGROUND

The U.S. Nuclear Regulatory Commission (NRC) is currently completing a test case modeling exercise to assist with the development and evaluation of guidance for performance assessment of LLW disposal systems. A summary of this modeling exercise was presented at this conference in 1994¹ and elsewhere.^{2,3} The test case involves a hypothetical, below-ground, concrete vault LLW disposal system located on a sub-humid coastal plain. The source term model¹ used for the test case considers the percolation of water into the concrete vaults and the release of radionuclides from the LLW waste by either rinse, diffusion, or dissolution mechanisms. The LLW waste form and inventory data were based on those from the "Richland '89" data.⁴

Because large amounts of cementitious material are present as various components of the disposal vault system, interactions between the infiltrating water and the concrete are expected to have a strong buffering effect on the chemical environment in and near the disposal vault system. An objective of our PNNL study was to assess suitable ranges of radionuclide solubility limits and distribution coefficients needed for calculating radionuclide release from the concrete-buffered chemical environment associated with disposal vaults considered in NRC LLW test case analysis. The radionuclides considered in our study included americium, inorganic carbon, chlorine, iodine, lanthanide elements, niobium, nickel, neptunium, plutonium, radium, strontium, technetium, thorium, and uranium. The scope of this paper is limited to our results for the solubility and adsorption of uranium. All of the results from our literature reviews and solubility calculations are described in

detail elsewhere.⁵

CHEMICAL ENVIRONMENT OF CEMENT/WATER INTERACTIONS

Cementitious materials have several important uses in LLW disposal facilities, including waste forms, backfill, and construction materials. Therefore, the long-term behavior of hydrated cements and their constituent phases in natural ground waters is important to understanding the potential release of radionuclides from LLW disposal systems. The composition of hydrated cement generally consists of 40-50 wt% calcium silicate hydrogel (C-H-S)⁸; 20-25 wt% portlandite [$\text{Ca}(\text{OH})_2$]; 10-20 wt% ettringite [$\text{Ca}_6\text{Al}_2\text{O}_6(\text{SO}_4)_3$], monosulfate [$\text{Ca}_4\text{Al}_2\text{O}_6\text{SO}_4$], and ferric phases; 10-20 wt% pore solution; 0-5 wt% minor components, such as NaOH, KOH, and $\text{Mg}(\text{OH})_2$.⁶ The chemical reactions associated with the hydration of cement are described in detail by others.^{7,8,9}

The composition of pore water that evolves during the degradation of cement in water has been studied extensively in the laboratory (see review in Krupka and Serne⁵). The dissolution of the C-H-S and portlandite phases, which may constitute as much as 75 wt% of the cement, have an important role in buffering the pH of the pore fluids. As ground water reacts with the cement, dissolution of the alkali hydroxide phases, present in relatively minor amounts, results initially in high pH values of approximately 13.5. As these phases are consumed in the cement, the pore fluid pH is then buffered near 12.5 by the dissolution of free portlandite in the cement. Eventually the portlandite is depleted and the pore fluid pH decreases to approximately 10.5 where it is controlled by the incongruent dissolution of the C-S-H hydrogel. When the dissolution of C-S-H is complete, the pH of the cement pore fluid will continue to decrease to a value buffered by the host ground water. This pH change will also be affected to a limited extent by the dissolution of any calcite that precipitated at the high pH conditions during the early stages of cement dissolution. The rate at which the pH of the pore solution changes from 13.5 to that of the ground water will depend on the physical properties (e.g., fractures, porosity, etc.) of the cementitious materials, rate of infiltration, chemistry of the infiltrating solution, and related hydrologic properties of the disposal system. For radioactive waste disposal systems being considered in the United Kingdom, studies¹⁰ indicate that the pH of the near-field pore water would remain above 10.5 for several hundred thousand years.

SOLUBILITY LIMITS

Conservative concentration limits based on equilibrium solubility constraints were calculated for several radionuclides for pore-water conditions considered in the NRC LLW performance assessment test case.⁵ The calculations are designed to address geochemical considerations that determine radionuclide solubility limits for the high pH conditions associated with cementitious systems.

Conceptual Model

The initial conditions for the geochemical evolution of the source term assumes that the composition of the leachate migrating from the disposal vault is controlled by the dissolution of the cement hydrate phases (i.e., "cement buffered" case) and characterized by pH values from 12.5 to 10.5. These conditions correspond to the initial stages of water infiltration into the LLW disposal system. In the final stages of the source-term evolution, the leachate composition is assumed to be controlled by reactions with soils and therefore equivalent to the local ground-water composition (i.e., "ground-water buffered" case). This case would correspond to an advanced state of degradation of the LLW disposal system when the availability of reactive concrete phases is insufficient to affect pore fluid compositions. The ground water infiltrating the disposal vaults has the following average composition in the test case: pH 5.8, Eh 500 mV, alkalinity (as CaCO_3) 14.1 ppm, Ba 0.2 ppm, Ca 4.0

ppm, Cl 3.0 ppm, F 0.07 ppm, Fe 0.118 ppm, K 1.3 ppm, Mg 0.4 ppm, Mn 0.052 ppm, (NO₃+NO₂) 0.6 ppm, Na 3.2 ppm, PO₄ 0.1 ppm, SO₄ 2.6 ppm, silica (as SiO₂) 5.8 ppm, and Sr 0.1 ppm.

The radionuclide concentration limits were calculated at fixed pH values at 0.5 pH-unit increments over the range of pH and redox (Eh) conditions defined by the cement buffered (pH = 12.5 and Eh = +200 mV) and ground-water buffered (pH = 5.8 and Eh = +500 mV) conditions. The Eh values used for the solubility calculations were assumed to vary linearly as a function of pH according to the equation: Eh (mV) = -44.78 pH + 760.

As a conservative limit, dissolved carbonate concentrations in the leachate were calculated between pH values of 5.8 and 8.0 by assuming equilibrium with 0.003 atm CO₂ (gas).¹¹ For pH values greater than 8.0, dissolved carbonate concentrations were calculated by assuming equilibrium with the solubility of calcite (CaCO₃). Calcite is known to precipitate as a result of reactions occurring with the dissolution of cement.^{12,13,14,15} The concentrations of dissolved calcium were also fixed in the pH range from 8.5 to 12.0 by assuming the leachate is equilibrium with the solubility of calcite. At pH 12.5, the leachate was assumed to be in equilibrium with the solubilities of both calcite and portlandite [Ca(OH)₂]. The solubility of portlandite is known to control the pH of hydrating cement at 12.5. To simplify the model calculations, the concentrations of the other dissolved, non-radionuclide constituents in the leachate were fixed at the concentrations specified for the ground-water composition listed. This assumes that the ground water is the major source for dissolved fluoride, chloride, nitrate/nitrite, sulfate, and phosphate. It should be noted that cement contains measurable quantities of sulfate and minor amounts of chloride phases that could increase the concentrations of these species in the pore fluids. Because the cement pore fluids at the outer edge of a vault will be diluted by infiltrating ground water, it was assumed that the leachates leaving the facility would have a moderate to low ionic strength.

MINTEQA2 Geochemical Model

The radionuclide solubilities and associated aqueous speciation equilibria were calculated using the chemical equilibria code MINTEQA2 (Version 3.11) obtained from the Center for Exposure Assessment Modeling at the U.S. Environmental Protection Agency (EPA) in Athens, Georgia. The MINTEQA2 code and its predecessor versions have been described by others.^{16,17,18,19} The MINTEQA2 calculations include aqueous speciation, solubility and saturation state (i.e., saturation index), adsorption, oxidation-reduction, gas phase equilibria, and precipitation/dissolution of solid phases. The thermodynamic database used to calculate radionuclide solubilities includes the EPA MINTEQA2 database augmented for aqueous species and solids containing the radionuclide elements of interest using database supplements provided by D. Turner [Center for Nuclear Waste Regulatory Analyses (CNWRA), Southwest Research Institute, San Antonio, Texas] and revisions added during the course of our study.

Solubility Limits for Dissolved Uranium

The concentration limits for the dissolved radionuclides were calculated using appropriate solubility-controlling solid(s) selected from those in the MINTEQA2 thermodynamic database. The solids were chosen based on phase-stability information given in published studies and knowledge of the geochemistry of radionuclide aqueous systems. Two solubility controls, schoepite [UO₂(OH)₂H₂O] and uranophane [Ca(H₃O)₂(UO₂)₂(SiO₄)₂·3H₂O], were considered in calculating concentration limits of dissolved uranium. Concentration limits based on schoepite are highly conservative. Schoepite is known to precipitate readily in low-temperature aqueous systems at laboratory time scales and result in high concentrations of dissolved uranium.²⁰ In natural low-temperature aqueous systems, the presence of alkali and/or alkaline earth ions at high pH conditions results in the precipitation of alkali/alkaline earth uranyl compounds that control the solubility of uranium at concentrations lower than those resulting from equilibrium with schoepite.

Uranophane is known to exist in uranium-loaded C-S-H gel mixtures and thus may be realistic solubility control for dissolved uranium. Calculation of its solubility, however, may be more susceptible to uncertainties in conceptual models and available thermodynamic data. Atkins et al.^{21,22} have investigated uranium interactions with $\text{Ca}(\text{OH})_2$ and C-S-H using a range of uranium loadings and equilibration periods of 21 to 75 days. Solid phases in the resulting mixtures were characterized by X-ray diffraction and analytical electron microscopy. Three uranium-containing phases were identified in these mixtures. These phases, which included uranophane, a hydrated calcium uranyl oxide [$\text{Ca}_2\text{UO}_5(1.2-1.5)\text{H}_2\text{O}$], and becquerelite [$\text{CaU}_6\text{O}_{19}\text{H}_2\text{O}$], could be possible solubility controls for uranium in cement-buffered systems. A source of thermodynamic data for the hydrated calcium uranyl oxide phase and becquerelite was not identified.

The concentrations calculated for dissolved uranium are listed in Table 1. The model results indicate that dissolved uranium would exist primarily in the +6 valence state, and uranyl carbonate and hydroxyl species would dominate the speciation of dissolved uranium at these pH/Eh conditions. At $\text{pH} > 10$, the hydrolysis species dominate the aqueous speciation of dissolved uranium. In addition to pH and Eh, the calculated solubilities of dissolved uranium are sensitive to the concentrations of dissolved carbonate as a result of strong complexation. Moreover, the concentrations of dissolved calcium and silica are additional factors affecting the uranium concentrations based on equilibrium with uranophane.

Table 1. Calculated Solubilities of Dissolved Uranium.

pH	Eh (mV)	Schoepite as Solubility Control	Uranophane as Solubility Control
-----mol/l-----			
5.8	500	1.5×10^{-5}	3.9×10^{-6}
6.5	470	3.9×10^{-5}	7.7×10^{-7}
7.0	450	1.1×10^{-4}	3.2×10^{-7}
7.5	420	3.9×10^{-4}	4.8×10^{-7}
8.0	400	1.1×10^{-3}	4.4×10^{-6}
8.5	380	2.9×10^{-4}	4.2×10^{-8}
9.0	360	2.6×10^{-4}	3.4×10^{-8}
9.5	340	2.6×10^{-4}	3.9×10^{-8}
10.0	310	2.8×10^{-4}	4.5×10^{-8}
10.5	290	3.8×10^{-4}	4.4×10^{-8}
11.0	270	7.8×10^{-4}	7.9×10^{-8}
11.5	250	2.3×10^{-3}	2.8×10^{-7}
12.0	220	7.5×10^{-3}	1.4×10^{-6}
12.5	200	3.0×10^{-2}	4.0×10^{-7}

Results of our solubility calculations bracket the experimental data reported in the literature. Ewart et al.²³ experimentally determined the concentrations of dissolved uranium resulting from the equilibration of cement-equilibrated waters that were oversaturated with dissolved uranium. The experiments involved the addition of sufficient U(VI) chloride solutions to cement equilibrated waters to obtain an initial concentration of 5×10^{-5} M dissolved uranium. The pH of the resulting mixtures was adjusted with sodium hydroxide or hydrochloric acid. Concentrations of dissolved

uranium predicted using the solubility of schoepite are several orders of magnitude greater than their experimental values. The solubility of uranophane, on the other hand, is in good agreement with the experimental values for pH values greater than 10.5, and significantly underestimates the concentrations at lower pH values. Ewart et al.²³ also modeled possible solubility controls for their experimental results, and noted that the solubility of schoepite overestimated uranium concentrations relative to their observed values.

RADIONUCLIDE SORPTION OF RADIONUCLIDES ON CEMENT/CONCRETE

The proper selection and use of distribution coefficients (K_d 's) that are most germane to the physicochemical system (e.g., cement-containing disposal vault) being modeled is an important concern to the technical defensibility of performance assessment calculations. The most common approach to quantifying adsorption is the use of K_d which is also often referred to as the distribution ratio, R_d . This constant is defined as the concentration of contaminant adsorbed on the solid per mass divided by the concentration of contaminant in solution per volume. The derivation and assumptions underlying the use of K_d in classical ion exchange literature have been discussed in detail by others.^{24,25}

Literature Review

We conducted a literature review of studies pertaining to radionuclide adsorption on cement and concrete materials. Our review of sorption information focused on the geochemical conditions associated with "fresh" cement/concrete where the cement pore waters will have $\text{pH} \geq 10$. Many factors⁵ lead to significant variability in reported K_d 's for the adsorption of radionuclides onto cement and concrete. Thus the proper selection of a K_d for a specific application will require judicious selections from the available literature. The adsorption measurements described in the literature are consistent on the qualitative adsorption (retardation) behavior of the radionuclides included in our review. That is, all sorption studies basically agree on which radionuclides are strongly retarded by cement paste (e.g., actinides, lanthanides, transition metals, inorganic carbon); moderately retarded by cement paste (e.g., radium, strontium, technetium); and not significantly retarded by cement paste (e.g., chlorine, cesium, iodine).

Tabulation of K_d 's for cementitious materials typically show considerable scatter (e.g., a factor of two difference in some studies) which could be misconstrued as inconsistency in sorption behavior. Because cement is usually studied in its freshly cured state, unusually large variations in reported K_d 's should be expected. At this point of the cement evolution, hydration reactions are continuing and alkali and OH^- ions are still leaching in significant quantities into the pore waters. Moreover, the resulting cement hydration minerals are not thermodynamically stable relative the geochemistry of most surface and ground waters.

From the perspective of hydrologic transport or performance assessment calculations, one must consider the fact that cement and concrete are a very impermeable yet very porous media. The major transport pathway through cement or concrete may in fact be through fractures or other defects that are more amenable to advective flow of water and more reactive to chemical weathering. The weathering products along these flow paths will exhibit different adsorption tendencies compared to the bulk cement hydration gel minerals. Several investigators^{26,27,28} have discussed the potentially beneficial phenomenon of carbonate plugging or armoring. Carbonate precipitation might close up fractures that form in monolithic cement and concrete structures and thereby further retard the already slow diffusion of most radionuclides through the hardened cement paste.

Categorization of K_d Values for Radionuclide Adsorption on Cement

The convention of Bradbury and Sarott²⁶ for the three physicochemical environments that all cements progress through was used to categorize K_d 's for radionuclide adsorption onto cement/concrete. These include the following: 1) *Environment I*, which occurs immediately after the cement hardens and is wetted by infiltrating water and where cement pore water is characterized as having a high pH of >12.5 , high ionic strength, and high concentrations of K and Na; 2) *Environment II*, where the soluble alkali salts are all dissolved and the cement pore water pH is controlled at 12.5 by the solubility of $\text{Ca}(\text{OH})_2$; and 3) *Environment III*, where the solubility C-S-H gel now controls the pH of the cement pore fluid, the ionic strength of the cement pore fluid is low, and its pH is ~ 10 or less. Each environment was split into an oxidizing and a reducing subenvironments, because some radionuclide elements may be present in chemically different forms depending on their redox-influenced valence state. We use the terms "oxidizing" and "reducing" to refer to the valence states of the specific radionuclide contaminant of interest.

The K_d 's of most radionuclides present in an Environment III cementitious system were assigned values of one-tenth of those K_d 's selected for radionuclides associated with Environment II following the convention of Bradbury and Sarott.²⁶ This assumption is necessary because laboratory data are not available for the sorption of radionuclides on severely weathered cements. Bradbury and Sarott reason that most of the very high surface area C-S-H gel has been removed in severely weathered cement. The remaining solid phase, silica, has significantly less surface area and adsorption capacity for trace contaminants compared to the C-S-H gel.

Adsorption of Uranium on Cementitious Materials

The Swedish studies^{29,30} present the most data for the adsorption of uranium onto cements or concretes. Uranium was added as the oxidized form U(VI) to the simulated cement pore waters. For the seven types of concretes and the weathered concretes, the K_d 's for uranium ranged from 350 to 13,000 ml/g. The average K_d was ~ 1000 , and the median value was 1400 ml/g. Although the behavior of uranium in aqueous systems is known to be sensitive to redox conditions, there was no significant increase in the uranium K_d measured in the experiments that used concrete containing blast furnace slag, an expected reductant.

Numerous investigators^{8,31,32} have studied the incorporation of U(VI) into cementitious waste forms. Under these conditions, the release of uranium is reduced significantly over the release of many other potentially soluble contaminants. Plausible solubility- and adsorption incorporation processes for U(VI) into the C-S-H gel are described by these investigators.

Based on the results of our review, it should be adequate to consider the adsorption of uranium, in either of its +6 or +4 valence states, as being readily retarded within the pore waters of cements and concretes where high pH conditions and relatively high concentrations of dissolved calcium exist. For this reason, the K_d 's selected for uranium on concrete and cement in Environments I and II are 1000 and 2000 ml/g for oxidizing and reducing conditions, respectively. For Environment III, assuming the one-tenth default convention, uranium K_d values of 100 and 200 ml/g are selected for oxidizing and reducing conditions, respectively.

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- a. Researchers in cement science use a convention to abbreviate names of cement phases. This terminology uses abbreviations for components in the cement phases, such as C = CaO, S = SiO_2 , A = Al_2O_3 , F = Fe_2O_3 , and, and H = H_2O . The formula of a cement phase is expressed as the appropriate stoichiometric combination of these initials, such as C_3AH_6 for $\text{CA}_3\text{Al}_2(\text{OH})_{12}$ [hydrogarnet].
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